Magnetic Excitations and Ordering in the Heavy-Electron Superconductor URu2Si2

Broholm, C.; Kjems, J.K.; Buyers, W.J.L.; Matthews, P.; Palstra, T.T.M.; Menovsky, A.A.; Mydosh, J.A.

Published in:
Physical Review Letters

DOI:
10.1103/PhysRevLett.58.1467

Magnetic Excitations and Ordering in the Heavy-Electron Superconductor URu_2Si_2

C. Broholm and J. K. Kjems
Risø National Laboratory, DK-4000 Roskilde, Denmark

W. J. L. Buyers and P. Matthews[(a)]
Atomic Energy of Canada Limited, Chalk River, Ontario, Canada K0J 1J0

and

T. T. M. Palstra, A. A. Menovsky,[(b)] and J. A. Mydosh
Kamerlingh Onnes Laboratorium, Rijksuniversiteit te Leiden, NL-2300 RA Leiden, The Netherlands
(Received 9 October 1986)

Neutron scattering demonstrates the coexistence of antiferromagnetic order and superconductivity below 1 K in the heavy-electron system URu_2Si_2. It is found that the upper (17.5 K) transition is to an antiferromagnetic phase with a (100) modulation wave vector and spins along the tetragonal c axis. The ordered moment is unusually small, $(0.03 \pm 0.01)\mu_B$. However, spin waves develop from damped, finite-gap excitations above T_N, and they are intense, propagating, and longitudinal with a zone-center gap of 1.8 meV.

PACS numbers: 75.30.Ds, 74.70.Tx, 75.25.+z

The relation between superconductivity and spin fluctuations has been a topic of intense interest since the discovery of heavy-electron superconductors such as UBe_{13}, UPt_3, and $CeCu_2Si_2$.[1] As in normal heavy-electron compounds these superconductors have large fluctuating localized moments at high temperatures and gigantic electronic specific-heat coefficients, $\gamma = Ce(T)/T$, at low temperatures. Several suggestions have been made that the superconductivity could be anisotropic and arise from the virtual attraction provided by the spin fluctuations rather than the s-wave pairing of the BCS phonon mechanism.[2] Neutron-scattering measurements on these systems have shown that spin fluctuations exist at low temperatures and that their spectra are continuous, extending to several tens of millielectronvolts.[3] This behavior is similar to that in many normal cerium and uranium compounds, where the presence of an Abrikosov-Suhl resonance near the Fermi energy leads to a large Kondo damping of the excitations of the f-electron system.[4]

Recently, a new heavy-electron compound, URu_2Si_2 ($\gamma = 180$ mJ/mole K^2) has been discovered[5,6] in which a lambda anomaly in its specific heat indicating a transition at 17.5 K is followed by the onset of superconductivity near 1 K. Walter *et al.*[7] have reported neutron-scattering experiments on a polycrystalline sample showing the existence of finite-gap magnetic excitations below T_N.

We have studied a single crystal of this compound by neutron scattering and will show that a weakly antiferromagnetic state develops below 17.5 K along with intense sharp spin-wave excitations characteristic of a localized f-electron system. Furthermore, the superconducting state develops from, and coexists with, this phase.

These results are important since they show that the superconducting pairs can be formed in an uniaxial antiferromagnet in which the low-lying excitations have the form of discrete spin waves. As in the heavy-electron superconductors the superconducting gap is found in a spectral region of very low density of spin fluctuations. It is low in UBe_{13}, and in UPt_3 because the broad paramagnon-type response peaks at large energies,[3] and in URu_2Si_2 because a spin-wave gap of 1.8 meV is formed. In all cases pairing caused by the spin-wave bosons remains a possibility; like phonons of the BCS theory their strength resides at energies much larger than the pairing energy and they are changed only slightly at the superconducting transition.

The present neutron-scattering experiments were performed at the cold-neutron source at the DR3 reactor of Risø National Laboratory and at the thermal-neutron source at the NRU reactor of the Chalk River Nuclear Laboratories.

The measurements were made on a 5-cm-long cylindrical URu_2Si_2 single crystal with a diameter of 0.5 cm, grown by a specially adapted Czochralski method.[8] The as-grown crystal was annealed for 7 days at 1000 °C. URu_2Si_2 has the body-centered tetragonal $ThCr_2Si_2$ structure with lattice parameters $a = 4.124$ Å, $c = 9.582$ Å at 4.2 K. The crystal had an a axis along the cylinder axis, and the measurements were performed with momentum transfers in the $(h0l)$ and $(hk0)$ planes.

The superconductivity was established in a separate experiment by ac-susceptibility measurements on two crystals cut from either end of the crystal used for the neutron-scattering experiment. The transition temperatures were $T_{c1} = 1.17$ K and $T_{c2} = 1.26$ K with a width between 10% and 90% of the transition $T_{c1} = 0.20$ K and $T_{c2} = 0.09$ K, respectively.

Figure 1 shows the temperature dependence of the in-

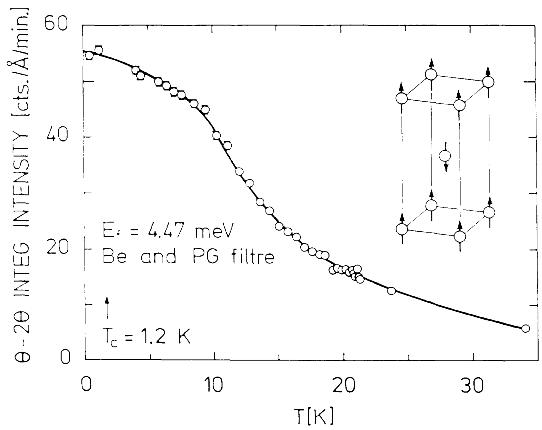

FIG. 1. Integrated elastic-magnetic Bragg scattering at (100) as a function of temperature, and the corresponding antiferromagnetic structure.

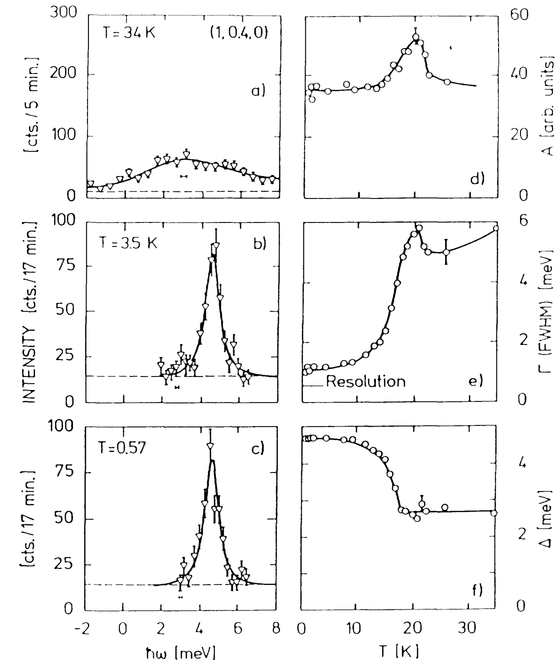

FIG. 2. Constant-q scans at $q=(1,0.4,0)$ in (a) the paramagnetic, (b) the antiferromagnetic, and in (c) the superconducting phase. The fixed final energy was 5 meV. (a) was measured with a horizontally focusing analyzer. In (b) and (c) the collimation was 60′-60′-53′-66′. The data taken with different analyzer configurations have been normalized, and plotted so equivalent cross sections correspond to the same height over the background. The solid lines in (a)-(c) are fits as described in the text, and (d)-(f) show the temperature dependence of the three parameters obtained from fits by the data similar to (a). The lines through these points are guides to the eye.

tegrated intensity at (100) which is a forbidden nuclear reflection. For this measurement the crystal was oriented with the c axis perpendicular to the scattering plane. Second-order contamination from the (200) nuclear reflection was suppressed with a beryllium and a pyrolytic graphite (PG) filter with use of an incident neutron wavelength of $\lambda=4.28$ Å. The deduced antiferromagnetic structure is shown in Fig. 1. The ordered moment is oriented along the tetragonal c axis since the magnetic Bragg intensities are found to be proportional to the polarization factor $(1-\hat{Q}_c^2)$, where $\hat{Q}_c$ is the c component of the unit vector parallel to the momentum transfer. Their intensities as a function of the magnitude of the momentum transfer Q decrease in approximate accordance with a squared U^{4+} form factor.[9] The scattering, therefore, arises from antiferromagnetically ordered $5f$-type moments.

The absolute scale of the ordered moment was determined by normalizing the integrated scattering to the weak nuclear (101) reflection. At 0.57 K the ordered moment is $(0.03\pm0.01)\mu_B$, an extremely low value compared to the high-temperature susceptibility along the tetragonal axis which is interpreted as an effective moment of $\mu_{eff}=3.51\mu_B$.[5] The (100) peak is not resolution limited, but has a Lorentzian line shape along C with a width of 0.018 Å^{-1} (FWHM). This width is significantly greater than the resolution at 0.0079 Å^{-1}, which was determined from a nuclear Bragg peak. The width along the a axis is 0.017 Å^{-1}, slightly larger than the resolution of 0.012 Å^{-1}. From these widths we estimate the range of the antiferromagnetic order to be about 100 interplanar spacings in the basal plane, but only 25 interplanar spacings along C. This may be related to crystallographic stacking faults in our crystal which were found to give rise to weak temperature-independent elastic scattering in rods along the c axis.

No other components of ordered moment were found in extensive single-crystal measurements at $T=10$ K on a four-circle diffractometer, nor in measurements on 13 g of URu_2Si_2 power. We can thus exclude the possibility of an unobserved ordered moment larger than $0.5\mu_B$, and we believe that we have found all of it.

As shown in Fig. 1 no anomaly in the ordered moment is observed at the superconducting phase transition. Antiferromagnetic order, therefore, coexists with superconductivity down to 0.57 K in URu_2Si_2.

We measured excitations below 10 meV at Risø National Laboratory with fixed scattered neutron energies of 5 meV or 14.5 meV with use of PG (002) as monochromator and analyzer. Be or PG filters were used to suppress higher-order contamination of the scattered beam. Excitations at higher energies were measured at Chalk River Nuclear Laboratories with fixed scattered neutron energies in the range 10-20 meV with use of a Si(111) monochromator and a PG (002) analyzer.

Figures 2(a)-2(c) show typical constant-q scans at different temperatures. Highly damped excitations

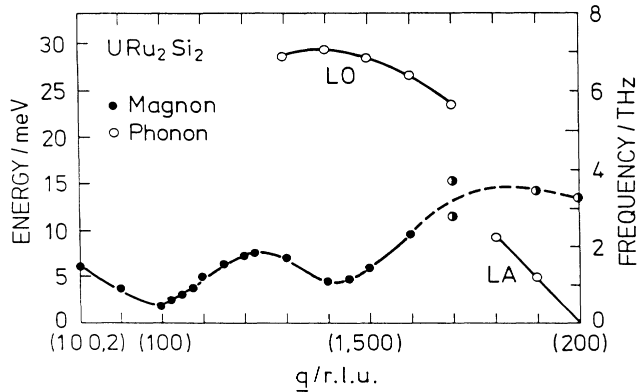

FIG. 3. Dispersion of magnetic (filled circles) and phonon (open circles) excitations along (ζ00) and partly (10ζ) at $T=4.2$ K. Half-filled circles denote a hybridized exciton-phonon mode. The lines are guides to the eye.

above T_N develop into sharp propagating magnons in the antiferromagnetically ordered phase. The dispersion of the modes at 4 K is shown in Fig. 3. The spin-wave cross section has the same dependence on momentum transfer and polarization factor as the ordered moment. This shows that the excitations belong to the $5f$-electron system and are polarized along the c axis. Sharp transverse magnetic excitations were searched for, but none was found up to 33 meV.

We have also measured the magnetic excitations in the superconducting phase at 0.57 K. Within the experimental uncertainty no change in intensity, energy, or damping relative to the measurements at 3.5 K occur, as shown in Figs. 2(b) and 2(c).

The sharpness of the observed magnetic excitations propagating along the a direction suggests that they occur within a single J multiplet. Their intensities are large. When normalized to an acoustic phonon, they correspond to a transition matrix element of $(2.2 \pm 0.2)\mu_B$. This large matrix element is in contrast to the extremely weak-ordered moment of only $(0.03 \pm 0.01)\mu_B$. The results suggest that URu_2Si_2 is a singlet ground-state system, with a weak induced moment. Since the spin wave is polarized along the ordered moment the excited state must also be a singlet. This is supported by the fact that the excitations do not split on the application of a magnetic field of 3 T along the c axis, although they do strengthen slightly. A singlet-singlet model is found to give a good description of the observed dispersion when long-range exchange coupling is included, and a singlet ground state is also qualitatively consistent with the low entropy change of $0.2R \ln 2$ observed through the 17.5-K transition.[5] The absence of transverse excitations below $\hbar\omega/k_B=400$ K also explains the axial anisotropy of the bulk susceptibility below 300 K. Finally, the singlet ground state along with the small ordered moment accounts for the finite susceptibility between T_c and T_N, the absence of a significant bulk susceptibility anomaly at T_N, and for the large amount of critical scattering above T_N.

One might thus attempt to describe the system assuming localized $5f^2$ states in the $^3H_4$ configuration on the U sites, and that the $J=4$ multiplet is split into five singlets and two doublets by a tetragonal crystal field.[10] However, this model fails to give a consistent description of the ordering temperature and the ordered moment, and of the q dependence of the spin-wave intensity. Furthermore, excitations with a propagation component, q_c, along the c axis show large damping, and as q_c is increased they merge into a continuum of magnetic scattering. These shortcomings of the simple localized crystal-field model indicate that it is necessary to take the strong hybridization with the conduction electrons into account more explicitly.[2,11-13]

Returning to the 17.5-K transition we note that thermal expansion measurements[14] show a minimum in the c/a ratio at 60 K which suggests that magnetoelastic effects are important as is often the case in singlet ground-state systems. This is further supported by high-pressure experiments.[15,16] Using a four-circle neutron diffractrometer, we looked for, but did not find, indications of lattice distortions and/or atomic rearrangements near T_N. Thermal expansion measurements show only small effects at T_N.[14] We conclude that the transition at 17 K is not a structural transition.

The most dramatic changes we have observed at T_N are in the excitation spectrum. Figure 2(a) shows a constant q scan at $T=34$ K with a momentum transfer corresponding to the minimum in the dispersion relation at (1,0.4,0). The evolution of this excitation into the ordered phase is summarized in Figs. 2(d)-2(f). Here we show the temperature dependence of the amplitude, A, damping Γ (FWHM), and first moment, Δ, of a resolution- and background-corrected Lorentzian response which describes our data well. Obviously, the phase transition is more clearly marked in the temperature dependence of Γ and Δ than in that of the ordered moment (Fig. 1). We suggest that the $5f$ electrons interact strongly with the gapless band electron system and that the dramatic change in the mode damping at T_N is due to a change in the low-energy electronic density of states consistent with the change in the electronic specific-heat coefficient, γ, at T_N. Indeed, the damping, Γ, of the magnetic excitations at (1,0.4,0) [Fig. 2(e)] and γ both decrease between 20 and 4 K by a factor of about 5.

In summary, we have shown that URu_2Si_2 is the first example of a heavy-electron system with propagating magnetic excitations and that these together with weak antiferromagnetic order coexist with superconductivity below 1.2 K. A significant part of the low-lying electronic density of states is removed at the antiferromagnetic transition leaving a gap in which the magnetic excitations can propagate. However, an approximate linear term remains in the specific heat just above the superconducting phase transition. This residual density of states, characterized by a γ of 40 mJ/mole K^2, is the heavy-electron Fermi liquid that ultimately becomes superconducting. The present results only allow us to speculate about the nature of the superconducting pairing, but the absence of shifts in the excitation frequencies on cooling below T_c does not rule out that the magnetic excitations provide the pairing boson for the electrons that remain, because the lowest excitation energy of 1.8 meV is much larger than the pairing energy. Furthermore, since the propagating magnetic excitations are longitudinal, it is expected from simple Fermi-liquid theory[17] that the pairing interaction for parallel spins is predominant. Hence an unconventional pairing state could be established below the magnetic transition by the propagating excitations. We are presently exploring this possibility with additional low-temperature neutron-scattering experiments.

We have enjoyed fruitful discussions with J. J. M. Franse, P. A. Lindgård, G. J. Nieuwenhuys, A. de Visser, and D. R. Taylor. Assistance by Dr. F. W. Poulson for grinding the polycrystalline sample is acknowledged. The work at Rijksuniversiteit te Leiden was supported by the Nederlands Stichting voor Fundamenteel Onderzoek der Materie (FOM).

[(a)]Permanent address: Queen's University, Kingston, Ontario, Canada K7L 3N6.

[(b)]Also at Natuurkundig Laboratorium der Universiteit van Amsterdam, NL-1018XE Amsterdam, The Netherlands.

[1]G. R. Stewart, Rev. Mod. Phys. **56**, 755 (1894).

[2]C. M. Varma, in *Moment Formation in Solids,* edited by W. J. L. Buyers (Plenum, New York, 1984), and Bull. Am. Phys. Soc. **29**, 404 (1984).

[3]G. Aeppli, E. Bucher, and G. Shirane, Phys. Rev. B **32**, 7579 (1985); A. I. Goldman, S. M. Shapiro, G. Shirane, J. L. Smith, and Z. Fisk, Phys. Rev. B **33**, 1627 (1986).

[4]W. J. L. Buyers and T. M. Holden, in *Handbook of the Physics and Chemistry of the Actinides,* edited by A. J. Freeman and G. H. Lander (North-Holland, Amsterdam, 1985), Vol. 2.

[5]T. T. M. Palstra, A. A. Menovsky, J. van den Berg, A. J. Dirkmaat, P. H. Kes, G. J. Nieuwenhuys, and J. A. Mydosh, Phys. Rev. Lett. **55**, 2727 (1985).

[6]W. Schlabitz, J. Baumann, B. Pollit, U. Rauchachwalbe, H. M. Mayer, U. Ahlheim, and C. D. Bredl, Z. Phys. B **62**, 171 (1986); M. B. Maple, J. W. Chen, Y. Dolichoouch, T. Kohara, C. Rossel, M. S. Torikachvili, M. W. McElfresh, and J. D. Thomson, Phys. Rev. Lett. **56**, 185 (1986).

[7]U. Walter, C.-K. Loong, M. Loewenhaupt, and W. Schlabitz, Phys. Rev. B **33**, 7875 (1986).

[8]A. A. Menovsky and J. J. M. Franse, J. Cryst. Growth **65**, 286 (1983).

[9]B. C. Frazer, G. Shirane, D. E. Cox, and C. E. Olsen, Phys. Rev. **140**, A1448 (1965).

[10]G. J. Nieuwenhuys, Phys. Rev. B (to be published).

[11]C. M. Varma, in *Theory of Heavy Fermions and Valence Fluctuations,* edited by T. Kasuya and T. Saso (Springer-Verlag, Berlin, Vienna, 1985), and in Comments Solid State Phys. **11**, 221 (1985).

[12]Y. Yafet, C. M. Varma, and B. Jones, Phys. Rev. B **32**, 360 (1985).

[13]A. C. Nunes, J. W. Rasul, and G. A. Gehring, J. Phys. C **19**, 1017 (1986).

[14]A. de Visser, F. E. Kayzel, A. A. Menovsky, J. J. M. Franse, J. van den Berg, and G. J. Nieuwenhuys, Phys. Rev. B **34**, 8168 (1986).

[15]F. R. De Boer, J. J. M. Franse, E. Louis, A. A. Menovsky, J. A. Mydosh, T. T. M. Palstra, U. Rauchswalbe, W. Schlabitz, F. Steglich, and A. de Visser, Physica (Amsterdam) **138B**, 1 (1986).

[16]M. W. McElfresh, J. D. Thompson, J. O. Willis, M. B. Maple, T. Kohara, and M. S. Torikachvili, Phys. Rev. B **35**, 43 (1987).

[17]M. T. Béal-Monod, C. Bourbonnais, and V. J. Emery, Phys. Rev. B **34**, 7716 (1986); P. W. Anderson, *Basic Notions in Condensed Matter Physics* (Benjamin, New York, 1985).